

Quantitative Two-Photon LIF Imaging of CO in Flickering CH₄/Air Diffusion Flames

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Introduction

Most detailed studies of chemical processes in diffusion flames have been carried out under steady flame conditions, which provide an experimentally reproducible environment for making careful profile measurements. In contrast, flickering diffusion flames exhibit a much wider range of time-dependent, vortex-flamesheet interactions, and thus they serve as an important testing ground for assessing the applicability of chemical models derived from steady flames to complex, turbulent flows. Two examples of particular interest are the production and oxidation of soot and CO. Both involve sufficiently slow chemical rates that one might expect to observe a strong sensitivity to the complex flowfields present in time-varying flames.

Measurements of soot concentrations in steady and moderately flickering CH₄/air diffusion flames burning with the same mean fuel flow rate have revealed a factor of 5 enhancement in the peak soot volume fraction and a factor of 4 increase in the time-averaged, volume-integrated soot levels for flickering flame conditions [1]. CO concentrations may also increase in the flickering flames, since carbon monoxide and soot compete for OH· and CO is the primary product of soot oxidation by OH· [2]. The goal of the present measurements is to quantify changes in the CO levels which occur in flickering diffusion flames, wherein increased soot production and subsequent oxidation may have important effects. For fuels more heavily sooting than methane (such as propane and ethylene), flickering flames are found to emit significant amounts of smoke for fuel flow rates which produce non-smoking steady flames [3]. For turbulent conditions, smoke and CO emission are expected to be closely linked.

Experimental Approach

Figure 1 shows a schematic diagram of the burner and phase-locked imaging setup, which have been described in detail previously [1,4]. Unconfined laminar diffusion flames were stabilized on a coannular burner with a 10.2 cm-diameter air annulus surrounding a 1.1 cm-diameter fuel tube, with a loudspeaker attached to the plenum below the fuel tube. The mean cold flow velocities for the methane fuel and the air coflow were 7.8 cm/s and 7.9 cm/s, respectively, for both the steady and flickering methane flames, giving a visible flame height of 79 mm. OH· concentrations have been previously measured in this steady CH₄ flame [5], and the soot properties have been extensively investigated as well [1].

Laser-induced fluorescence measurements of CO were performed in the steady flame and in both moderately and strongly flickering flames produced by applying sine waves of magnitude 0.75 V and 1.5 V (peak-to-peak) to the plenum loudspeaker. As with previous studies [1,4], the optical diagnostics were phase-locked to a sinusoidal oscillation of the fuel stream at the 10 Hz repetition rate of a Nd³⁺:YAG laser, permitting phase-specific measurements to be performed in the time-varying flowfields. This frequency is close to the natural flame flicker frequency caused by buoyancy-induced instabilities (~12 Hz). In contrast to our prior experiments, a synthesized function generator replaced both a variable delay pulse generator and a signal generator to provide exact reproducibility of the flickering flame forcing amplitude and the detection phase settings. Figure 2 illustrates OH· fluorescence and scattering images for the moderately flickering condition.

Two-photon laser-induced fluorescence from CO was observed by exciting the Q branch (0,0) band of the B¹Σ⁺ - X¹Σ⁺ transition with linearly polarized light at 230 nm. Fluorescence from the B¹Σ⁺ - A¹Π (0,1) band was detected at 483.5 nm. This excitation and detection scheme has been implemented for flame measurements by

Haumann et al. [6], Seitzman et al. [7], Tjossem and Smyth [8], and Smyth and Tjossem [9]. The incoming light had a pulse energy of 0.8-1.3 mJ and was focussed at the burner centerline to a beam diameter of 440 μm (FWHM) using a 500-mm focal length lens. The fluorescence signal was recorded at 90° to the direction of propagation of the laser light as a line image on an intensified CCD camera using an 18 ns collection gate and a narrow-band dielectric filter with a bandpass of 479.8 ± 5.0 nm. For optimum detection sensitivity an f/1.4 collection lens was used.

Diagnostic Issues

In the flickering methane flames the maximum soot volume fraction reaches a level of 1.5-2.0 ppm in the clipped portions of the flame. Severe interferences are observed upon excitation with ultraviolet light from these relatively high soot concentrations and the associated molecular precursors of soot. Diagnostic issues of interest include the following:

1. The intensity levels required for two-photon excitation of CO also lead to broadband laser-induced incandescence from soot particles, broadband fluorescence due to large molecular species (attributed to polycyclic aromatic hydrocarbons, PAH), and laser-induced production of C₂ Swan band emission. As a result, profile measurements must be taken using narrow-band detection to minimize these interferences. The interference signals exhibit different power dependences, which necessitates close attention to the laser fluence. Net CO profiles are obtained tuning the laser wavelength on and off resonance and subtracting the measured signals.
2. The temperature range where significant CO levels are expected is relatively narrow, 1500-2100 K, in these axisymmetric co-flowing diffusion flames, but the temperature field has not been measured for the flickering flame conditions. Modeling of the Q-branch rotational spectrum indicates that selection of the $J'' = 32$ rotational line yields a mole fraction measurement that is least sensitive to the temperature variations in this range. However, the relatively weak fluorescence signal from this high J line combined with intense background interferences necessitates selection of a stronger (lower J'') transition. Measurements were carried out using $J'' = 20$, for which the largest mole fraction errors are +38% at 1500 K and -41% at 2100 K when calibrating the CO concentration at 1800 K. Comparison of area-integrated profiles at a given height above the burner, with versus without temperature corrections, gives much smaller errors, typically 1-5%.
3. The laser intensities needed for successful imaging experiments are sufficiently high that photoionization dominates all loss processes from the excited $B^1\Sigma^+$ state [8]. This situation leads to several advantages: (1) no quenching corrections are required, (2) the CO fluorescence is linear with laser fluence, making laser intensity corrections straightforward, and (3) the emission wavelength can be tuned to match the transmission curve of a specific narrow-band filter and also avoid detection of C₂ (1,0) Swan band emission by proper selection of the initial rotational line.

Results

Figure 3 presents CO concentration profiles for the steady methane flame and one phase at moderate forcing conditions for which tip clip-off occurs. Maximum CO concentrations are approximately equal for the steady and flickering conditions, in agreement with the recent tunable diode laser measurements of CO in the same flames by Skaggs and Miller [10]. For flickering flames, the greater radial extent of the burning flamelet following clip-off yields larger area-integrated CO levels. Overall, however, the increased soot production in the flickering flames coupled with longer residence times [11] does not lead to appreciable increases in CO levels.

CO imaging measurements were also attempted for more heavily sooting ethylene flames, in which interferences from laser-induced soot incandescence and broadband fluorescence are severe. Only in regions in the steady flame where $f_v \leq 5$ ppm could CO concentration data be obtained, and measurements in flickering ethylene flames proved to be intractable.

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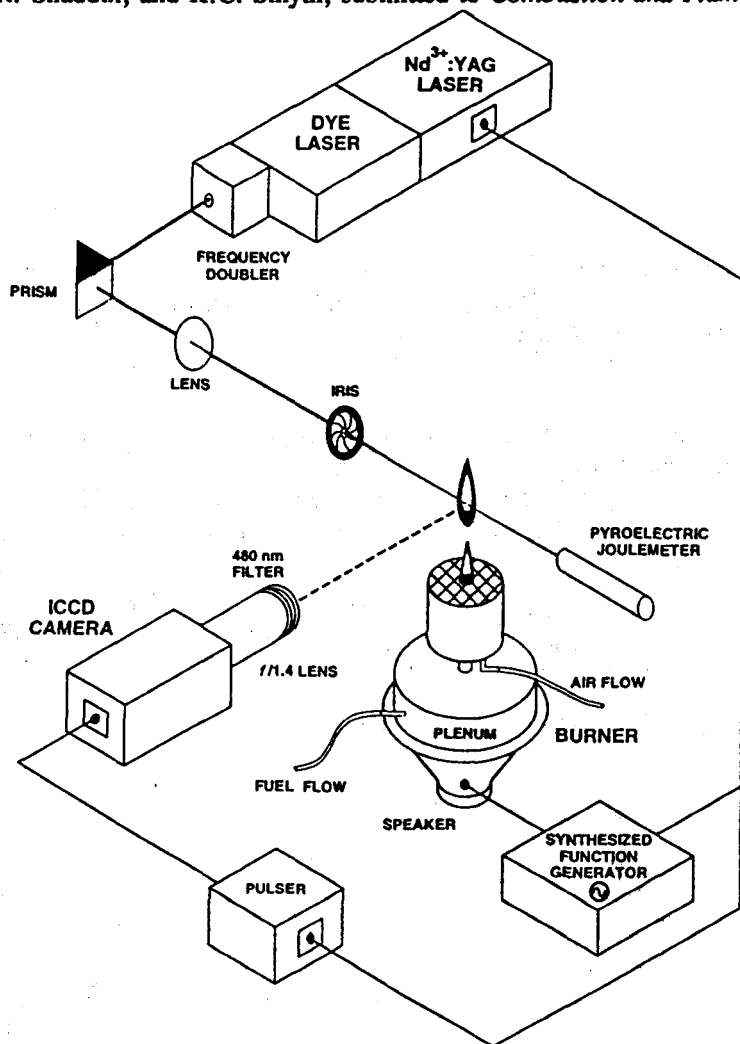


Figure 1. Experimental set-up for 1-D imaging of axisymmetric diffusion flames which are acoustically excited and phase-locked to the pulsed dye laser system operating at 10 Hz. Images are recorded using an intensified charge-coupled device (ICCD) camera. The coannular burner is mounted on a vertical translation stage, allowing measurements to be performed from the burner lip up to a height of 11.5 cm.

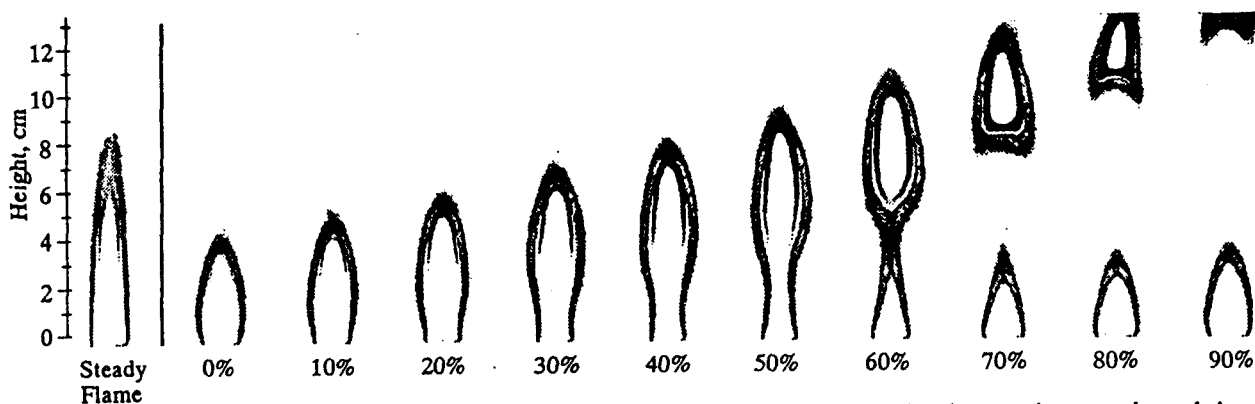


Figure 2. Laser energy-corrected OH· laser-induced fluorescence and soot scattering images in a steady and time-varying laminar CH₄/air diffusion flame using horizontally polarized light at 283.55 nm. The visible flame height of the steady flame is 79 mm above the fuel tube exit. For the flickering flame, a 0.75 V loudspeaker excitation is employed; ten phase increments separated by 10 ms are shown with an arbitrary zero phase.

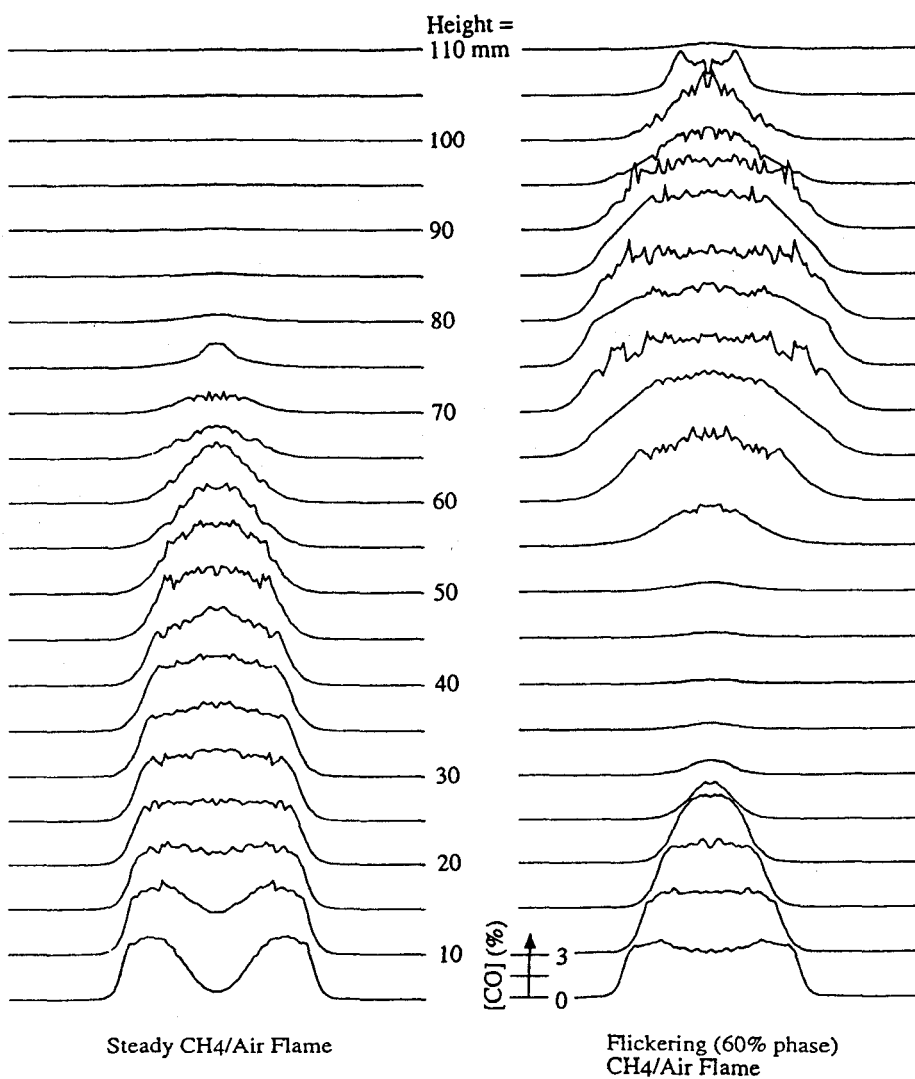


Figure 3. Quantitative CO concentration profiles for the steady CH₄/air diffusion flame and the moderately flickering flame at 60% phase (see above).